

EXAMINER'S AMENDMENT

An examiner's amendment to the record appears below. Should the changes and/or additions be unacceptable to applicant, an amendment may be filed as provided by 37 CFR 1.312. To ensure consideration of such an amendment, it MUST be submitted no later than the payment of the issue fee.

Authorization for this examiner's amendment was given in a telephone interview with Catherine L. Bell on July 15, 2008.

Page 2 of amendment, dated July 8, 2008

Claim 1, line 1 replace "to produce" with "for preparing a"

Page 3

Claim 1, line 16 replace "one of R⁴ and R⁷ are" with "one of R⁴ and R⁷ is"

Page 4

Claim 4, line 1 replace "copolymer" with "process"

Claim 5, line 1 replace "copolymer" with "process"

Claim 8, line 1 replace "copolymer" with "process"

Claim 9, line 1 replace "copolymer" with "process"

Page 5

Claim 24, line 1 replace "propylene polymer" with "process"

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Claim 30, line 5 replace “ 5 ” with “ 5 ” (no italics)

Page 6

Claim 30, line 16 replace “one of R⁴ and R⁷ are” with “one of R⁴ and R⁷ is”

Page 7

Claim 30, line 7 delete “about” which appears prior to the term “28 wt %”

Claim 30, line 7 insert “of ethylene derived units” after “28 wt %”

Claim 31, line 1 replace “copolymer” with “process”

Claim 31, line 10 replace “analogs” with “analogues”

Page 9

Claim 32, line 9 replace “one of R⁴ and R⁷ are” with “one of R⁴ and R⁷ is”

Claim 32, line 13 delete “about” which appears prior to the term “28 wt %”

Page 10

Claim 34, line 1 delete “, when present,”

Claim 36, line 2 delete “, when present,”

Page 12

Claim 40, line 14 delete “, when present,”

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Page 13

Claim 40, line 2 delete “about” which appears prior to the term “28 wt %”

Claim 43, line 1 delete “, when present,”

Page 15

Claim 48, line 13 delete “, when present”

Page 16

Claim 48, line 3 delete “about” which appears prior to the term “28 wt %”

Claim 48, line 3 insert “units derived from” after the term “28 wt %”

Claim 51, line 1 delete “, when present,”

Page 18

Claim 56, line 12 replace “one of R⁴ and R⁷ are” with “one of R⁴ and R⁷ is”

Claim 56, line 17 delete “about” which appears prior to the term “28 wt %”

Page 19

Claim 57, line 1 delete “, when present,”

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Basis for amendments:

Claim 1 replacing “to produce” with “for preparing a”
language of all independent claims is internally consistent

Claims 1, 30, 32, and 56

replace “one of R⁴ and R⁷ are” with “one of R⁴ and R⁷ is”
subject/verb agreement

Claims 4, 5, 8, 9, and 31

replace “copolymer” with “process”
independent claim 1 is drawn to a process, preamble of dependent claims
changed accordingly

Claims 24 replace “propylene polymer” with “process”
independent claim 1 is drawn to a process, preamble of dependent claim
changed accordingly

Claim 30 replace “ 5 ” with “ 5 ”
remove italics; matter of form rather than substantive change

Claims 30, 32, 40, 48, and 56

delete “about” which appears prior to the term “28 wt %”
specification does not provide support for the endpoint “about 28 wt %”

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Claim 30 insert "ethylene derived units" after "28 wt %"
ethylene comonomer is defined in definition of copolymer

Claim 31 replace "analog" with "analogues"
spelling correction

Claims 34, 36, 40, 43, 48, 51, and 57

delete "when present"
structural elements are present in claimed compounds, therefore, conditional term
"when present" is not applicable

Claim 48 insert "units derived from" after "28 wt %"
copolymer contains units derived from ethylene rather than ethylene monomer

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Allowable Subject Matter

The following is an examiner's statement of reasons for allowance: Claims 1, 4, 5, 8-10, 24, and 30-63 are allowed over the closest references cited below.

The present invention is drawn to a process for preparing a propylene copolymer in the presence of specified catalyst wherein the copolymer exhibits the property $g'_{88-89}/g'_{20-60} \geq 1.10$ wherein subscripts 88-98 and 20-60 refer to the weight percent of copolymer eluted in GPC-DRI and g' is the weight average g' over the elution range designated by subscripts 88-98 and 20-60. The term g' is an average index and represents the ratio of intrinsic viscosity of the copolymer to the intrinsic viscosity of a reference polymer. The claimed ratio of g' values implies that the intrinsic viscosity of the higher weight fraction is at least 1.1 times greater than the intrinsic viscosity of the lower weight fraction. This reflects an increasing comonomer distribution at higher molecular weight. This characteristic is exhibited in inventive copolymers having a comonomer content of 5-28 wt % (see data in Table 3).

Claims 1, 4, 5, 8, 9, 10, 24, and 31 are drawn to a process for preparing propylene copolymer comprising polymerizing propylene and ethylene in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 10-20 wt % of ethylene derived units, M_w of 20,000-1,000,000 and exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.20$. The metallocene compound is a bridged, *bis*-(indenyl) hafnocene (M = Hf) complex; salient features include: a carbon-based bridging group having general formula $-C(R^{14})(R^{15})-$, $-(R^{14})(R^{15})C-C(R^{14})(R^{15})-$, *inter alia*, and at least one of substituents R^4 and R^7 , located on the 4- and 7-position of the indenyl ligand, respectively, is not hydrogen (see claims for full structural details).

Claim 30 is drawn to a process for preparing propylene copolymer composition comprising polymerizing propylene and ethylene in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 5-28 wt % of ethylene derived units, M_w of 20,000-1,000,000 and

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exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$. The metallocene compound is a bridged, *bis*(indenyl) metallocene complex; salient features include: M is a group 4, 5, or 6 metal, a carbon-based bridging group having general formula $-C(R^{14})(R^{15})-$, $-(R^{14})(R^{15})C-C(R^{14})(R^{15})-$, or $-(R^{14})(R^{15})C-C(R^{16})_2-$ wherein R^{14} , R^{15} , and R^{16} are identical and are a C_1 - C_4 alkyl group, and at least one of substituents R^4 and R^7 is not hydrogen (see claims for full structural details).

Claims 32-39 are drawn to a process for preparing propylene copolymer comprising polymerizing propylene and a comonomer selected from ethylene and C_4 - C_{10} in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 5-28 wt % of the comonomer, M_w of 20,000-1,000,000 and exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$. The metallocene compound is a bridged, *bis*(indenyl) hafnocene complex; salient features include: a carbon-based bridging group having general formula $-(CR^9R^{10})_m-R^8-(CR^9R^{10})_n-$ wherein $m = 1$ or 2 and $n = 1$ or 2 , and at least one of substituents R^4 and R^7 is not hydrogen (see claims for full structural details).

Claims 40-47 are drawn to a process for preparing propylene copolymer comprising polymerizing propylene and a comonomer selected from ethylene and C_4 - C_{10} in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 5-28 wt % of the comonomer, M_w of 20,000-1,000,000 and exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$. The metallocene compound is a bridged, *bis*(indenyl) hafnocene complex; salient features include: a carbon-based bridging group having general formula $-(CR^9R^{10})_m-R^8-(CR^9R^{10})_n-$ wherein $m = 1$ or 2 and $n = 1$ or 2 , and substituents R^4 and R^7 are identical and are one of a fluorine, a chlorine, a bromine, a C_1 - C_4 alkyl group, or a C_6 - C_{10} aryl group (see claims for full structural details).

Claims 48-55 are drawn to a process for preparing propylene copolymer comprising polymerizing propylene and ethylene in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 5-28 wt % of units derived from ethylene, M_w of 20,000-1,000,000 and exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$. The metallocene compound is a bridged, *bis*(indenyl) hafnocene

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complex; salient features include: a carbon-based bridging group having general formula $-(CR^9R^{10})_m-R^8-(CR^9R^{10})_n-$ wherein $m = 1$ or 2 and $n = 1$ or 2 , and substituents R^4 and R^7 are identical and are one of a fluorine, a chlorine, a bromine, a C_1 - C_4 alkyl group, or a C_6 - C_{10} aryl group (see claims for full structural details).

Claims 56-63 are drawn to a process for preparing propylene copolymer comprising polymerizing propylene and a comonomer selected from ethylene and C_4 - C_{10} in the presence of the product of an activator and a metallocene compound wherein the propylene copolymer has at least 50 wt % of units derived from propylene, 5-28 wt % of the comonomer, M_w of 20,000-1,000,000 and exhibits a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$. The metallocene compound is a bridged, *bis*(indenyl) hafnocene complex; salient features include: a carbon-based bridging group having general formula $-(CR^9R^{10})_m-R^8-(CR^9R^{10})_n-$ wherein $m = 1$ or 2 and $n = 1$ or 2 , substituents R^2 and R^3 , located on the 2- and 3-positions of the indenyl ligand, respectively, are hydrogen, and at least one of substituents R^4 and R^7 is not hydrogen (see claims for full structural details).

Winter *et al.* (U.S. 5,679,811) discloses a catalyst system for polymerization of olefins wherein the catalyst comprises the reaction product of aluminoxane and a silicon or germanium bridged, *bis*(indenyl)hafnocene complex. The reference teaches that catalysts of silicon or germanium bridged *bis*(indenyl)hafnocene complexes afford higher molecular weight polymer compared with corresponding ethylene bridged *bis*(indenyl)hafnocenes. Clearly, the reference does not teach or fairly suggest preparation of catalysts containing metallocenes of the instant claims, and hence, the reference can not teach the process of the instant claims.

Winter *et al.* (U.S. 5,304,614) teaches a catalyst system for polymerization of olefins wherein the catalyst comprises the reaction product of activator and a bridged, *bis*(indenyl) zirconocene or hafnocene complex in which the 4- and 7-positions of the indenyl ligand are not hydrogen; see general formula (Ia). The inventors disclose the surprising result that propylene and higher 1-olefin are polymerized to give polymers of relatively low molecular weight, but the behavior of inventive metallocenes toward ethylene is completely different; very high molecular weight polyethylene is prepared with excellent catalyst activity. Representative examples show that catalysts prepared from Me_2Si and Et bridged zirconocenes having 4-/7-substitution afford

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propylene homopolymer having weight average molecular weight ranging from 8000 to 16,000. Including a methyl substituent in the 2-position (*i.e.*, $\text{Et}(2,4,7\text{-Me}_3\text{Ind})_2\text{ZrCl}_2$) results in considerable increase in propylene homopolymer molecular weight (30,6000 and 61,100). In one example, a catalyst derived from $\text{Et}(4,7\text{-Me}_2\text{Ind})_2\text{ZrCl}_2$ produces polypropylene homopolymer having Mw of 32,000 (reaction temperature at 30 °C). Propylene copolymer a prepared from a catalyst derived from $\text{Me}_2\text{Si}(4,7\text{-Me}_2\text{Ind})_2\text{ZrCl}_2$ produces having an ethylene content of 3.1 wt % is prepared having molecular weight of only 15,600. Although generic formula (Ia) encompasses Me_2C bridged metallocenes, metallocenes containing more than two contiguous atoms in the bridging group, and hafnium analogues of the zirconocenes shown in the examples, the reference, taken as a whole, does not teach or suggest a process for preparing propylene copolymer having least 50 wt % of units derived from propylene, 5-28 wt % of comonomer, M_w of 20,000-1,000,000 and exhibiting a ratio $g'_{88-89}/g'_{20-60} \geq 1.10$ in the presence of a catalyst prepared from the claimed metallocenes. Therefore, the subject matter of the instant claims is patentably distinct over Winter *et al.*

Winter *et al.* (DE 38 25 814) discloses a process for polymerization of polyolefin homopolymer or copolymer in the presence of a catalyst comprising a bridged, *bis*(indenyl) hafnocene complex, represented as generic formula (I). The reference does not teach use of complexes having a particular substitution pattern on the indenyl ligand, and there is little guidance with respect to a process of making a particular copolymer other than indication that propylene copolymer may be prepared having up to 50 wt % of ethylene as comonomer. Working examples disclose preparation of propylene homopolymer in the presence of a catalyst derived from $\text{Et}(\text{Ind})_2\text{ZrCl}_2$. The reference, taken as a whole, does not teach or suggest a process for preparing the claimed propylene copolymer in the presence of a catalyst prepared from the claimed metallocenes. Therefore, the subject matter of the instant claims is patentably distinct over Winter *et al.*

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Imuta *et al.* (U.S. 5,504,172) discloses catalysts prepared from a bridged, *bis*(indenyl) metallocene complex in which each indenyl ligand is substituted in the 4- and 7-positions with an alkyl group and wherein the bridging group may be a divalent hydrocarbon group containing up to four contiguous carbon atoms. Inventive catalysts are useful for preparing propylene-ethylene copolymer having an ethylene content of 0.5 to 5 mole %. In an alternative embodiment, the propylene elastomer comprises 50-95 mole % of propylene units, 5-50 mole % of ethylene units, and not more than 10 mole % of units other than propylene and ethylene. Working examples show preparation of propylene-ethylene copolymer from Me_2Si or Ph_2Si bridged zirconocenes; ethylene content in product is variable and depends on ligand substitution pattern, nature of the bridging group, and reaction temperature. The reference, taken as a whole, does not teach or suggest a process for preparing the claimed propylene copolymer in the presence of a catalyst prepared from the claimed metallocenes. Therefore, the subject matter of the instant claims is patentably distinct over Imuta *et al.*

Schottek *et al.* (EP 1 052 263) discloses catalyst prepared from a series of group 4 *bis*(indenyl) metallocene complexes wherein indenyl ligands are linked by a single carbon atom (*i.e.*, bridging group is $-\text{CH}_2-$, $-\text{CMe}_2-$, $-\text{CPh}_2-$, $-\text{CEt}_2-$, $-\text{C(H)(Me)-}$, $-\text{C(H)(Et)-}$, $-\text{C(Me)(Et)-}$) and wherein the indenyl ligand is substituted in the 2-position with an alkyl group and in the 4-position with an alkyl or substituted aryl group. A single hafnocene complex $\text{H}_2\text{C(2-Me-4-}i\text{-BuPh-Ind)}_2\text{HfCl}_2$ is described. Catalysts are useful for preparation of a variety of polymer products, and there is mentioned ethylene-propylene copolymer. Working examples disclose preparation of polypropylene homopolymer with bridged zirconocene complexes. The reference, taken as a whole, does not teach or suggest a process for preparing the claimed propylene copolymer. Therefore, the subject matter of the instant claims is patentably distinct over Schottek *et al.*

Sachetti *et al.* (U.S. 6,399,533) discloses preparation of HPDE and LLDPE in the presence of catalyst derived from alkylene bridged, *bis*(indenyl) metallocenes. The reference does not teach or suggest preparation of the claimed propylene copolymer.

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Sadatoshi *et al.* (U.S. 5,830,968) discloses propylene random copolymer containing a propylene component and a C₄-C₁₀ α -olefin component wherein the α -olefin content lies in the range of 6-40 wt %. Ethylene may also be copolymerized during preparation of the copolymer as long as the ethylene does not impair the desirable physical properties of the copolymer. Inventive copolymer is prepared from a catalyst derived from Et(Ind)₂ZrCl₂, although inventors contemplate use of Et(4-MeInd)₂HfCl₂, Et(7-MeInd)₂HfCl₂, and Et(4,7-Me₂Ind)₂HfCl₂ as catalyst component. The reference, taken as a whole, does not teach or suggest a process for preparing the claimed propylene copolymer. Therefore, the subject matter of the instant claims is patentably distinct over Sadatoshi *et al.*

Machida *et al.* (U.S. 2004/0106738) discloses a catalyst comprised of Et(4,7-*i*Pr₂Ind)₂HfCl₂ and Me₂Si(benzInd)₂ZrCl₂ for preparation of propylene homopolymer in a first stage and followed by polymerization of propylene in the presence of homopolymer from the first stage and 1,9-decadiene. Inventive polymers exhibit a branching index *g* in the range of $0.75 \leq g < 1.0$ in the molecular weight range of $2 \times 10^6 - 10 \times 10^6$ and branching index in the range of $0.90 \leq g \leq 1.0$ in the molecular weight range of 500,000 - 2×10^6 . The reference does not disclose the process recited in the instant claims.

Tsutsui *et al.* (U.S. 5,336,746) discloses preparation of random copolymers of propylene having from 1-10 mole % of a C₄-C₂₀ α -olefin in the presence of a catalyst comprising a hafnium complex having two cycloalkadienyl groups linked together *via* an alkylene linkage. A generic formula is not provided, however, the metallocenes Et(4-MeInd)₂HfCl₂, Et(7-MeInd)₂HfCl₂, and Et(4,7-Me₂Ind)₂HfCl₂ are listed as useful for practicing the invention. Polymers containing 90-99 mole % of propylene units, 0.5-9.5 mole % ethylene, and 0.5-9.5 mole % of C₄-C₂₀ α -olefin units are also prepared with inventive catalysts (col. 6, lines 30-37). An ethylene content of about 9.5 mole % equates an ethylene content of less than 8 wt %. Tsutsui *et al.* does not teach preparation of polymer of the instant claims, which contains 10-20 wt % of ethylene derived units. Tsutsui *et al.* does not elucidate fully the scope of the term "alkylene bridge" and all hafnocene complexes listed in the patent contain the unexceptional ethylene bridging group. Based on these facts, it is deemed the reference does not disclose or suggest adequately non-

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ethylene-bridged metallocenes recited in the instant claims, and one of ordinary skill in the art would not have sufficient motivation to modify the structural bridge of compounds listed in the prior art. As such, the prior art does not teach or suggest a process for preparing the claimed propylene copolymer, and thus, the claimed invention is patentably distinct over Tsutsui *et al.*

Göres *et al.* (U.S. 6,350,830) teaches a supported catalyst comprising a support, a cocatalyst, and a *rac/meso* isomer mixture of a group 4 *bis*(indenyl) metallocene represented by general formula (I) wherein each indenyl ligand is substituted in the 4-position with substituted or unsubstituted phenyl group. Alkylene-based bridging groups are selected from Et, -Me₂C-CH₂-, -MeCH-CHMe-, -Me₂C-CMe₂-, Me₂C, Ph(Me)C, or Ph₂C. Specific hafnocenes include Et(2-Et-4-*i*-BuPhInd)₂HfBz₂, Et(2-Et-4-*i*-BuPhInd)₂HfMe₂, and Et(2-Et-4-*i*-BuPhInd)₂Hf(Me₂)₂. Inventive catalysts find use in a process for preparation of polyolefin homopolymer or copolymer, with preference given to copolymerization of propylene with ethylene and/or with one or more 1-olefin having from 4 to 20 carbon atoms and/or one or more dienes having from 4 to 20 carbon atoms. The prior art does not provide adequate suggestion to make propylene-ethylene copolymer having the claimed comonomer content. Moreover, catalysts of Göres *et al.* are supported, and these represent a different class of catalyst than that of Applicant (polymers of the instant invention are prepared in solution phase using non-supported catalyst). Given the unpredictable nature of catalysts, and in light of the fact that catalysts of the prior art are fundamentally different from those of the instant invention, one having ordinary skill in the art would not reasonably expect to make the claimed propylene copolymer from the catalyst of Göres *et al.* The reference, taken as a whole, does not teach or suggest a process for preparing the claimed propylene-ethylene copolymer, and therefore, the subject matter of the instant claims is patentably distinct over Göres *et al.*

Any comments considered necessary by applicant must be submitted no later than the payment of the issue fee and, to avoid processing delays, should preferably accompany the issue fee. Such submissions should be clearly labeled "Comments on Statement of Reasons for Allowance."

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Any inquiry concerning this communication or earlier communications from the examiner should be directed to Rip A. Lee whose telephone number is (571)272-1104. The examiner can be reached on Monday through Friday from 9:00 AM - 5:00 PM.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Vasu S. Jagannathan, can be reached at (571)272-1119. The fax phone number for the organization where this application or proceeding is assigned is (571)273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <<http://pair-direct.uspto.gov>>. Should you have questions on the access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll free).

/Rip A. Lee/
Art Unit 1796

July 15, 2008

/VASUDEVAN S. JAGANNATHAN/
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